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Work on this project is performed in close connection between ASU, DCA, and J.A. Woollam Co. in the developing of real time in-situ thickness control project. ASU has become a testbed in developing of ultra-stable manipulator for MBE chamber by DCA and for Woollam Co. for their developing of 44 wavelength ellipsometer in real time control. Researcher at ASU include Prof. G. N. Maracas, Dr. Chau-Hong Kuo, and Mr. Martin Boonzaayer. Tapani Levola from DCA and Blain Johs from Woollam Co. was also involved in the construction and testing of the working equipment and model for this project. The results and experience that we learned from this project can be used not only in MBE growth, but it can also be applied in the MOCVD chamber which is widely used in the industry for the thin film deposition. ARPA can transfer this technique from the university funded project to industry into various MOCVD chamber to grow thin film quantum device. The quality of the thick layer of SiO<sub>2</sub> grown on various substrate, like glass or plastic, have been measured ex-situ by ellipsometry. This result has been used to monitor the quality of SiO<sub>2</sub> thin film on plastic or glass. This report will mainly concentrated in the accomplishment at the last year of this contract.

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# Final Technical Report

July 1, 1992 - December 31, 1995

## "In-situ Growth Monitoring of Molecular Beam Epitaxy Processes.

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## 1. Introduction

This is the final technical progress report on ARPA/ONR contract N00014-92-J-1931 titled "In-situ Growth Monitoring of Molecular Beam Epitaxy Processes." The progress at the end of the third year is summarized in the following area.

- \* Finished measurement and analysis of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  optical constant,  $x$  range from 0.1 to 1.0 and increment of 0.1, with temperature range from 27 °C to 625 °C with 25 °C increment.

- \* Construction(with DCA) and testing of a mechanically stable MBE manipulator for in-situ optical measurement and this is commercial available to MBE chamber from various vender through DCA.

- \* Testing of 44 wavelength spectroscopic ellipsometer, constructed by J.A. Woollam Co.), for tracking MBE layer thickness during growth.

- \* Test 44 wavelength ellipsometer for reproducibility in real time in-situ thickness control in MBE chamber by growing Fabry-Perot cavity. The AIAs and GaAs optical constants at growth temperature obtained from this contract was used as database to monitor thickness of AIAs and GaAs layers during growth.

Work on this project is performed in close connection between ASU, DCA, and J.A. Woollam Co. in the developing of real time in-situ thickness control project. ASU has become a testbed in developing of ultra-stable manipulator for MBE chamber by DCA and for Woollam Co. for their developing of 44 wavelength ellipsometer in real time control. Researcher at ASU include Prof. G. N. Maracas, Dr. Chau-Hong Kuo, and Mr. Martin Boonzaayer. Tapani Levola from DCA and Blain Johs from Woollam Co. was also involved in the construction and testing of the working equipment and model for this project. The results and experience that we learned from this project can be used not only in MBE growth, but it be can also applied in the MOCVD chamber which is widely used in the industry for the thin film deposition. ARPA can transfer this technique from the university funded project to industry into various MOCVD chamber to grow thin film quantum device. The quality of the thick layer of  $\text{SiO}_2$  grown on various substrate, like glass or plastic, have been measured ex-situ by ellipsometry. This result has been used to monitor the quality of  $\text{SiO}_2$  thin film on plastic or glass. This report will mainly concentrated in the accomplishment at the last year of this contract.

## 2. Experimental

### 2.1 $\text{Al}_x\text{Ga}_{1-x}\text{As}$ optical constants

A simple two phase model (vacuum/thin film) was used to determine the dielectric constants of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  directly from SE data. The detailed of the data analysis for these data can be found elsewhere.[1] Figure 1 shows a plot of the imaginary part of the dielectric function for  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  ( $x=0.214$ ) for various temperature ranging from room temperature to approximately 600 °C. The two peaks at 3.062 and 3267 eV correspond to the  $E_1$  and  $E_1+\Delta_1$  inter band transitions at room temperature. The higher energy peaks are the  $E_0'$  and  $E_2$

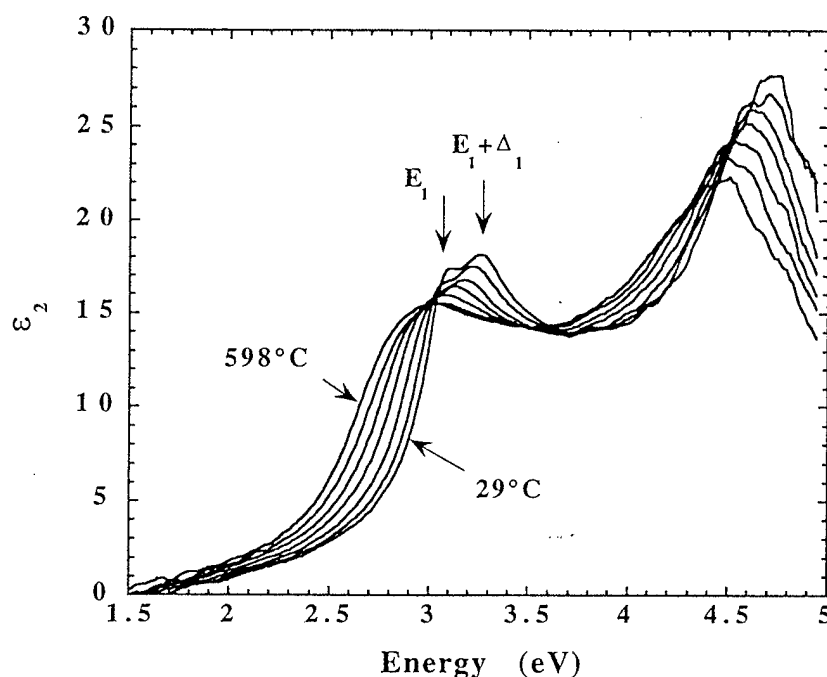


Fig. 1 Imaginary part of the dielectric function for AlGaAs with Al composition of 21.4% at temperature of 29, 106, 212, 312, 420, 519, and 598 °C.

transitions. From Fig. 1, as the substrate temperature increases, the inter band transitions decrease in energy as expected. Also, the amplitude of all the amplitude decrease with temperature increase. This trend can be explained by temperature broadening for the inter band transitions. For higher Al compositions the aforementioned trends are also present. Fig. 2 shows a similar plot of  $\epsilon_2$  for an Al composition of 0.74.

For AIAs we determined that the  $\epsilon_2$  value for the as-grown 1  $\mu\text{m}$  thick layer were lower than expected in the short wavelength ( $<5000\text{\AA}$ ). Another indication is the oscillation part of the delta value from SE data never reach value of 0. All the evidence above indicated a degradation of the surface during growth. It was reported that the growth of AIAs by MBE on GaAs(100) at 600 °C resulted in rough surface is because of the low diffusion length of Al on the growing surface.[2] Consequently, a two step method was adopted to grow AIAs for the measurement presented here. A 1  $\mu\text{m}$  thick of AIAs was grown at 600 °C then annealed at 700 °C for 30 minutes. Further annealing produce no change in the value of the ellipsometric parameters. To investigate the cause of the discrepancy between the as-grown and annealed AIAs layer, the optical constants derived from the annealed layer was used to fit the as-grown data.

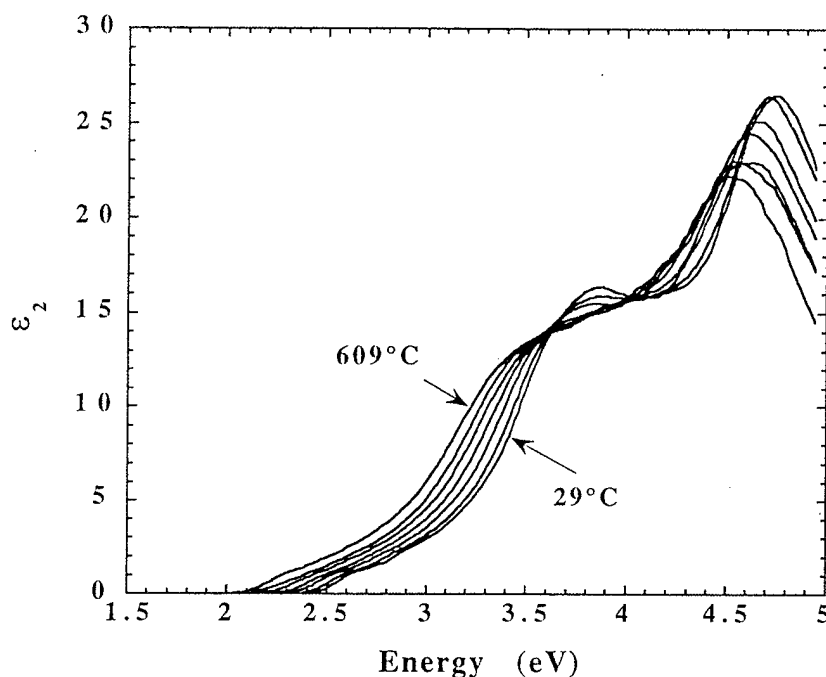


Fig. 2 Imaginary part of the dielectric function measurement for AlGaAs with Al composition of 74 % at temperatures of 29, 109, 213, 321, 426, 508, and 609 °C.

Excellent agreement was found when the surface roughness was introduced in the model. (The ellipsometric data was further supported by HRTEM for as-grown layer and annealed layer.) As a result, caution must be exercised when using AIAs optical constants for real time thickness control of epitaxial growth.

## 2.2. Ultra-stable manipulator

The schematic of DCA MBE growth chamber with 44 wavelength ellipsometer is shown in Fig. 3. As shown in the diagram, part of the supporting rod in the manipulator was cut away and substitute with pizeo crystal. Due to the outgasing of cement used in pizeo crystal during the bake out of growth chamber, the pizeo crystals were contain inside vacuum seal bellows. The length of each supporting rod can be adjusted from the high voltage supply outside of the growth chamber with the maximum adjustment of 200  $\mu\text{m}$ . The wobbling of the substrate surface during substrate rotation is coming from two different parts. One is coming from the difference in length for three supporting rods. This can be corrected by machining the supporting rod until the problem is fixed before the installation of manipulator. The other problem is the substrate holder. For different substrate holder, there is a difference in flatness for each substrate holder. We can adjust the length of the supporting rods to fit one

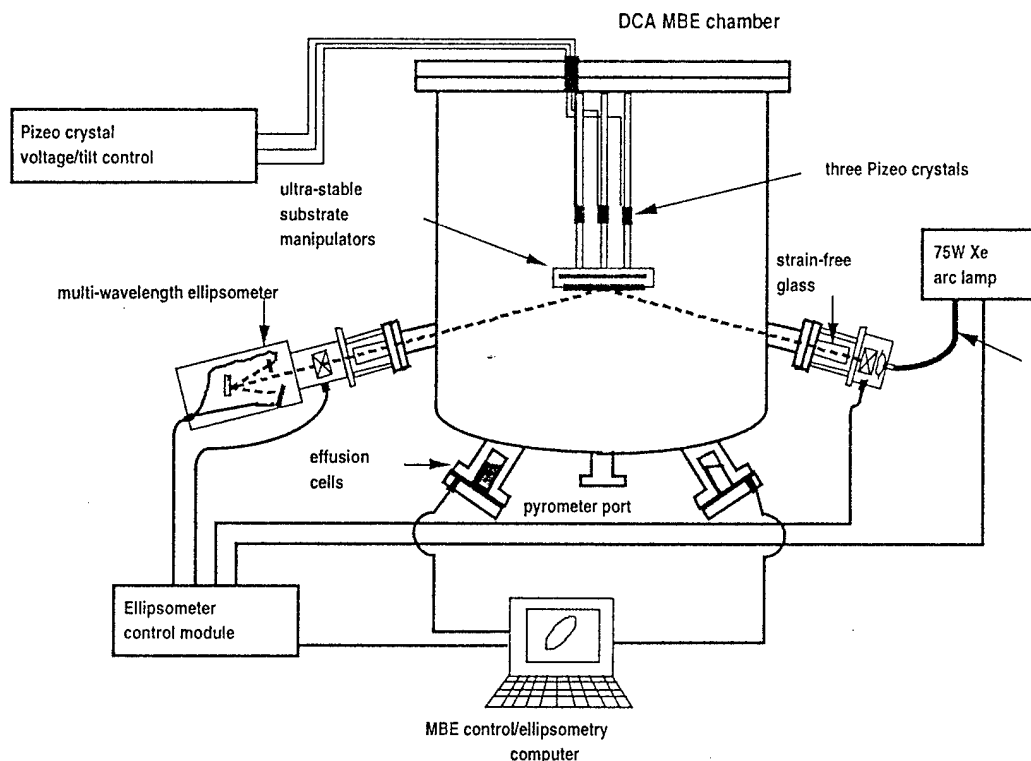


Fig. 3 Schematic of DCA MBE vacuum chamber.

substrate holder at one particular orientation to load the wafer, but we can not use another substrate holder in the experiment.

The wobbling of the substrate surface during the substrate rotation can be monitored from the 44 wavelength ellipsometer. A quad detectors was located at the entrance of the 44 wavelength entrance slit. Deviation of the reflected light from substrate surface away from the center of the entrance slit will cause a difference of the signal from quad detectors. The wobbling of the substrate surface from rotation was determined to be  $\pm 0.2^\circ$ . The wobbling normally cause a fluctuation in ellipsometric data and hence increase the error of the determination of ellipsometric data. In some extreme case, the reflected light is not entering the detector slit and the ellipsometric data is lost. By adjusting high voltage supply to each pizeo crystal, the signal from quad detectors will show a sigh of reducing wobbling. The wobbling of the substrate was reduced by factor of 10 and the wobbling is determined to be  $\pm 0.02^\circ$  which is better than any other existing manipulator design. We can use any substrate holder in the lab. for the in-situ experiment without worry about wobbling problem anymore.

### 2.3. In-situ real time thickness control

Growth of Fabry-Perot cavities used for vertical cavity surface emitting lasers (VCSEL) and electro-optic modulator was used to demonstrate in-situ real time MWE thickness control. Such structures consist of a cavity having an optical mode placed in between two dielectric mirrors. This particular structure

was used to demonstrate the MWE control of thick epitaxial layers because the position of the FP mode is very sensitive to the change of the cavity thickness. Distributed Bragg reflectors (DBR) consisting of alternating  $1/4\lambda$  high and low index of refraction layers produces mirrors that have a high reflectance wavelength band. High reflectivity indicates good thickness control of the DBR layers.

The FP cavity designed for this experiment consisted of 10 period of top and 10.5 periods of bottom DBRs having AlAs/GaAs thickness of 822.2 Å and 679.4 Å respectively. The GaAs  $1\lambda$  FP cavity was designed to have a mode at 970nm so the thickness of the GaAs was nominally 2717.5 Å.

To analyze ellipsometric data collected from MWE, the optical constants of AlAs and GaAs at growth temperature, 600 °C, are needed. The bulk material file of GaAs and AlAs [3,4] from our previous studies was used in this experiment to obtain thickness information in real time. Another important aspect of real time control is to reduce the time required to obtain thickness information for a complicated structure like DBR. The virtual interface model was used to reduce the data analysis to a simple three phase model (vacuum / epitaxial / substrate).

Six samples of  $1\lambda$  FP cavities were grown to demonstrate the reproducibility of the MWE thickness control. Sample 1 to sample 4 were grown under the same Al and Ga cell temperature conditions over the period of one month. The same Ga and Al cell temperatures were used for all four different samples. The variation of FP mode from four different samples was less than 2.1 nm from the normal incidence reflectance measurement. This indicates a difference of less than 0.2 % in thickness control of the  $1\lambda$  GaAs cavity. The FP mode measured at the edge of the 2 inch wafer has a blue shift of 5 nm compared to the FP mode at the center of the 2 inch wafer. This indicates the layer thickness uniformity across the 2 inch wafer is about 0.5 %.

Sample 5 was grown with perturbed Al and Ga cell temperatures during the growth of AlAs and GaAs layers. Since the thickness control program is monitoring epitaxial layer thickness information in real time, we do not expect the flux fluctuation to affect epitaxial layer thickness control. As shown in Fig. 4, the FP mode from sample 5 stay at the center of variation from the early four samples. This indicates that constant calibration of growth rate for binary epitaxial layer is not necessary in MWE thickness control experiment.

Sample 6 was grown with Si doping,  $1 \times 10^{18}$ , in all the epitaxial layers. Since virtual interface model and bulk material optical constants are used in analyzing thickness information, a constant surface temperature is assumed during the growth of epitaxial layers. The effect of the increasing surface temperature while Si shutter open and possible change of the epitaxial layer optical constants due to Si doping do not take into account in data analysis. However from sample 6 FP mode as shown in Fig. 4, the difference between sample 6 and the average value from sample 1 to 4 is less than 0.1 %. The

effect of surface temperature changing and Si doping can be neglected in in-situ DBR thickness control experiment.

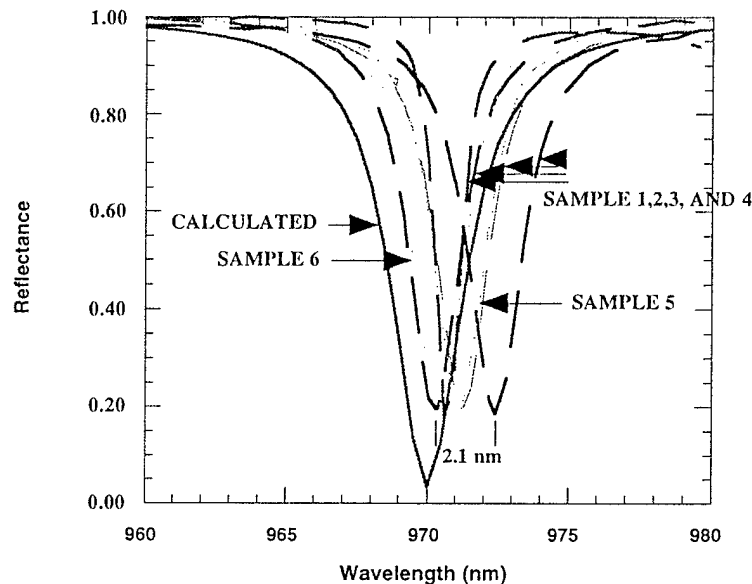


Fig. 4. Normal incidence reflectance of a 970 nm (design) FP cavity. Superimposed on the calculation is the measured curve showing FP mode from different samples.

From this experiment we demonstrate the reproducibility of the in-situ ellipsometric thickness is quite good. With the help of the recent progress in personal computer. It is possible that the throughput of the thickness information, which include data collection and data analysis, can be reduced to 100 ms or less instead of 3 second at the present time. It is foreseeable that ellipsometry technique can be applied to MOCVD, which has a growth rate 10 times higher than typical MBE. Since MOCVD is widely used in the industry, this will be a great improvement for the industry to improve their quality control of thin film growth.

#### 2.4. Ex-situ measurement of SiO<sub>2</sub> thin film growth on plastic

This is a project in connection with Professor Thomas Sigmon's graduate student Dave Shea. The main purpose is to determine the quality of the SiO<sub>2</sub> thin film growth on the plastic without destroy the thin film. Since the preparation of the plastic surface before thin film growth is critical to the quality of the SiO<sub>2</sub> thin film. It is important to apply non-destructive method like ellipsometry to have a fast feedback which is less than 10 minutes. The information obtained from this can be used to improve either the surface preparation of plastic or to adjust growth parameters of SiO<sub>2</sub>.



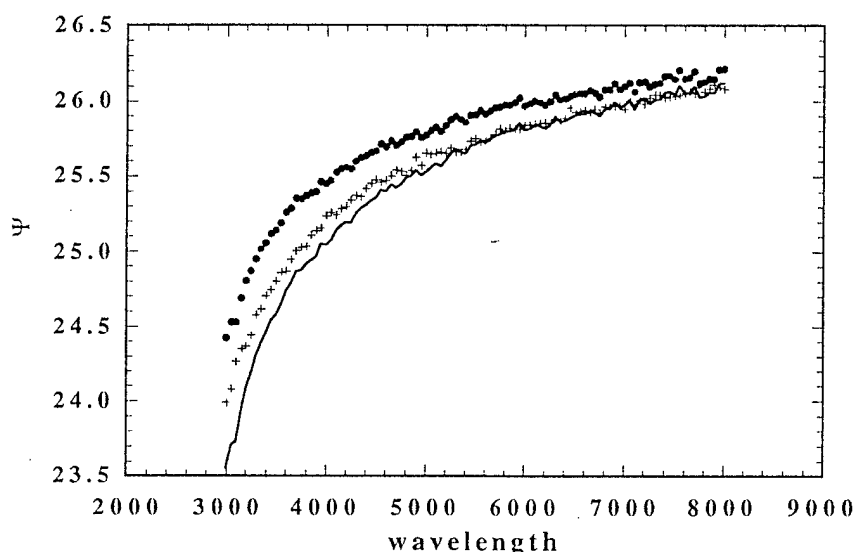


Fig. 5 Comparison of measured ellipsometric data  $\Psi$  from model. Solid line is the measured  $\Psi$  value, + is from EMA model which is the composition of  $\text{SiO}_2$  thin film with void, and • is from model with  $\text{SiO}_2$  thin film only.

There is no optical constants information available for plastic in visible light range to analyze  $\text{SiO}_2$  thin film. The ellipsometric data was analyzed with the simple two phase model (vacuum / substrate). In order to remove the interference from the plastic's backside reflection, the backside of the plastic was scratched with #100 sandpaper. The optical constants of the plastic used in this experiment is a direct calculation from ellipsometric measurement.

The ellipsometric data of plastic with thin film was analyzed with three phase model (vacuum / thin film / plastic). The ellipsometric data were obtained from Spectroscopic ellipsometry. In order to obtain a best fit value from the model, the thin film in the model has to be substituted with EMA layer which is a composition of  $\text{SiO}_2$  and void. The best fit value for the model from EMA layer is 88.4 % void with 344 Å layer thickness. The best fit value for  $\text{SiO}_2$  only thin film is 114 Å for layer thickness. The EMA approximation give a better fit between model and measured value as shown in Fig. 5. This indicates that only 11.6 % of the area has  $\text{SiO}_2$  thin film growth. This result was latter confirmed by SEM with Au coating on  $\text{SiO}_2$  film. This demonstrated that ellipsometry can also be used ex-situ to optimize thin film growth condition without destroy thin film surface as SEM did.

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